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abstract

A critical examination of the spin dynamics in high- $T_C$  cuprates is made on the light of recent inelastic neutron scattering results obtained by different groups. The neutron data show that incommensurate magnetic peaks in YBCO belong to the same excitation as the resonance peak observed at  $(\pi/a, \pi/a)$ . Being only observed in the superconducting state, the incommensurability is then rather difficult to reconcile with a stripe picture. We also discuss the link between the resonance peak spectral weight and the superconducting condensation energy.

After more than ten years of intense investigations, the precise role of antiferromagnetic (AF) correlations for the mechanism of the high-temperature superconductivity remains a puzzling and open question. Since the early days, it has been obvious that both phenomena are clearly connected just by looking at the generic phase diagram of high- $T_C$  cuprates. Of course, a competitive role rather than cooperative between long-range antiferromagnetism and superconductivity was generally inferred as both phenomena are thought to occur in exclusion of each other. The next key question was then: are the dynamical AF correlations observed in the superconducting (SC) range of the phase diagram prejudicial or responsible for superconductivity ?

A necessary step to put some insight into this still unsolved question is the knowledge of the spectral weight of the spin susceptibility,  $\chi(Q, \hbar\omega)$ .  $\chi(Q, \hbar\omega)$  would, for instance, enter the SC pairing interactions in any mechanism based on antiferromagnetism. As a matter of fact, Inelastic Neutron Scattering (INS) is the only technique which directly measures the full energy and momentum dependences of the imaginary part of the spin susceptibility. Further, the amplitude of  $Im\chi(Q, \hbar\omega)$  can be determined in absolute units by a calibration of the magnetic neutron intensity versus other scattering such as phonons. This has been done only recently for the high- $T_C$  cuprates and, brings essential insight for the relation between AF correlations and superconductivity as we shall see below.

This technique is limited by the need of large single crystals (of  $\text{cm}^3$  size) usually difficult to grow in complex systems such as high- $T_C$  cuprates. This has reduced the number of systems which could be studied to a very few:  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO),  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  (YBCO) and only recently  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (BSCO). Further, INS spectra can be sometimes ambiguous to analyze as, for instance, neutron scattering also directly measures the phonon spectrum which is typically of the same order of magnitude. Of course, a lot of effort has been developed to overcome these difficulties. However, this situation has postponed the emergence of a full agreement between the different groups. However, clear unmistakable features have been established which have considerable impact on the role of AF fluctuations. Here, we shall emphasize

some key aspects on the basis of published data by the different groups. It should be mentioned that as far as the raw data are concerned, a fairly good agreement can be noticed. Disagreements are rather related to the data analysis which sometimes leads to clearly different conclusions.

## The resonance peak: a collective spin excitation of *d*-wave superconductors

Among the observed magnetic features [1], the "AF resonance peak" observed below  $T_C$  is certainly one of most important results which has been widely studied since its discovery in 1991 by Rossat-Mignod *et al* [2] in YBCO<sub>6.92</sub>. When entering the SC state and **only** below  $T_C$ , a sharp (almost energy resolution limited) spin excitation appears in the neutron scattering data at an energy,  $E_r$  and at the AF wavevector  $Q_{AF} = (\pi/a, \pi/a)$  ( $Q_{AF}$  is the propagation wavevector of the AF state of the insulating undoped parent compound,  $a=3.85$  Å is the 2D square lattice parameter).

The striking characteristic of the resonance peak is actually its temperature dependence. Indeed, its energy,  $E_r$ , does not shift towards lower energy when approaching  $T_C$  (a shift of at most  $\sim 4$  % can be inferred [3, 4, 5]) but its intensity is vanishing upon heating at the superconducting temperature  $T_C$  for all doping levels, actually following an order parameter-like behavior. Recently, an attempt has been made[6] to associate the vanishing of the resonance peak intensity in underdoped sample with the temperature  $T^*$  where the resistivity displays the so-called "pseudo-gap" anomaly. This statement is not correct being based on an arbitrary analysis. Indeed, neither the data published by Dai *et al* [5, 6] nor our own data [7, 8, 9] provide any justification for a separation in the normal state (NS) of the spectrum into resonant and nonresonant parts. No published temperature dependence of the neutron intensity at the resonance energy (or more correctly, at the energy transfer where the resonance peak appears in the SC state) suggests an anomaly at a temperature  $T^*$  larger than  $T_C$ . A clear upturn is systematically observed only at the SC transition temperature. In our opinion, this incorrect attribution of the "onset of the resonance peak at  $T^*$ " has been made from the fact that the broad maximum of the spin susceptibility in the normal state occurs in some underdoped sample roughly at the same energy as the resonance peak [1, 9]. But, as a matter of fact, the apparent equivalence of the normal state energy and the resonance peak energy breaks down in underdoped samples closer to optimal doping [1, 10].

Interestingly, the resonance energy scales with the SC temperature as:  $E_r \sim 5.2k_B T_C$ . This relation holds in the two systems where the resonance peak has been observed so far, YBCO[1, 9] and BSCO[11]. This actually is not only valid at optimal doping but also remains correct on both sides of the high  $T_C$  phase diagram: on the underdoped side, as experimentally realized for different oxygen contents in YBCO[1, 9], as well as on the overdoped side as observed in a BSCO sample[12]. This generic relationship of  $T_C$  with the temperature-independent resonance energy calls for an explanation which is not obvious when one considers the different models usually invoked to interpret the resonance peak (See Refs [1, 9, 12] for a discussion of these approaches).

Further, the resonance feature appears to be strongly sensitive to parameters which affect the superconducting properties. For instance, the substitution of Zn impurities within the CuO<sub>2</sub> plane in YBCO, known to strongly reduce the SC temperature ( $dT_C/dy \simeq -12$  K/%) [13] without changing the doping level[14], induces a rapid vanishing of the resonance intensity: small amounts of zinc impurities ( $y$  ranging from 0.5% to 2% in YBa<sub>2</sub>(Cu<sub>1-y</sub>Zn<sub>y</sub>)<sub>3</sub>O<sub>6+x</sub>)[15, 16, 17] are sufficient to remove its spec-

tral weight without strong renormalization of the resonance energy itself. In contrast, magnetic Ni impurities which are three times less efficient to remove superconductivity ( $dT_C/dy \simeq -4$  K/%) [13], have also less effect on the resonance peak intensity and keep the ratio  $E_r/k_B T_C$  almost unchanged [17]. This extreme sensitivity of the resonance feature to defects affecting the SC transition temperature then might explain why no resonance peak has been reported so far in the LSCO system whose maximum  $T_C$  ( $\sim 40$  K) is anomalously low as compared to other single  $\text{CuO}_2$  layer systems where  $T_C$  can reach 90 K (Tl- or Hg- based system). The disorder which might be responsible for the reduction of  $T_C$  in LSCO can also remove the resonance peak feature. Further, Zn and Ni impurities in YBCO [16, 17] also produce a systematic broadening in energy of the resonance peak, by  $\sim 10$  meV. Similar broadening found in BSCO [11, 12] can then be naturally accounted for by the presence of intrinsic defects in that system.

Until recently, the resonance peak has been widely described as a single commensurate excitation. Although this statement remains certainly correct in the slightly overdoped YBCO<sub>7</sub> system, we have recently demonstrated [18] in YBCO<sub>6.85</sub> that the resonance peak actually exhibits a full dispersion curve away from  $(\pi/a, \pi/a)$  momentum. This illustrates, on experimental grounds, that the resonance peak can be considered as a collective mode of the superconducting state of high- $T_C$  cuprates as theoretically proposed (see e.g. [19, 20, 21]). The observed downward dispersion actually relates the commensurate resonance peak with the incommensurate peaks observed at lower energy and recently reported in underdoped YBCO [22, 23]. By detailed temperature dependences of the neutron intensity at different wavevectors and energies, we have established [18] a dispersion compatible with the following relationship,

$$E_r(q) = \sqrt{E_r^2(Q_{AF}) - (\alpha q)^2} \quad (1)$$

where  $q$  is the wavevector measured from  $Q_{AF} = (\pi/a, \pi/a)$ .  $E_r(Q_{AF}) = 41$  meV is the previous commensurate resonance energy, and  $\alpha \simeq 125$  meV.Å represents an isotropic dispersion relation. Certainly, the relation Eq. 1 is only a first approximation which needs to be refined. Indeed, the measured wavevector pattern at a fixed energy  $E = 35$  meV located below  $E_r(Q_{AF})$  exhibits an intensity modulation in the 2D  $(H, K)$  momentum space shown in Fig. 1 with larger intensity in the directions (100) or (010) and lower intensity in the directions (110) or  $(1\bar{1}0)$ . Such detailed momentum dependence (which reproduces the shape reported in YBCO<sub>6.6</sub> [22] at 24.5 meV (below  $E_r(Q_{AF}) = 34$  meV) as well as that discussed in [23]), implies a modification in the dispersion relation of Eq. 1. For instance, an anisotropy of  $\alpha$  between the (100) and (110) directions should be added and would certainly account for the momentum pattern of the neutron intensity shown in Fig. 1. Although the resonance peak dispersion is, so far, only evidenced in one sample, YBCO<sub>6.85</sub>, we think it is a generic feature of the spin dynamics in the superconducting state over a wide part of the high- $T_C$  cuprate phase diagram. Data reported in Refs. [22, 23] are fully consistent with such an interpretation although this has not been discussed this way. For sure, more work is needed to generalize this conclusion, for instance, to give the actual doping dependence of the  $\alpha$  parameter.

The observation of incommensurate peaks [22, 23], in addition to the commensurate resonance peak, has stimulated several theoretical models in Fermi liquid-like theories. It has been discussed as a combined effect of both i) topology of the band structure and ii) anisotropic superconducting order parameter either at the level of the bare susceptibility [24, 25] or after taking into account of the interactions by a random phase approximation [21, 26, 27]. Furthermore, a dispersive collective mode has been predicted to arise below the particle-hole spin-flip continuum in the  $d$ -wave super-

conducting state as a result of a momentum-dependent pole in the spin susceptibility pulled by antiferromagnetic interactions [20]. Our recent observation of a downward dispersion [18] supports the latter proposal. However, to fully establish the collective nature of the resonance peak, a necessary step will be to observe the particle-hole spin-flip continuum. In any case, our recent data demonstrate that superconductivity affects not only the energy lineshape of the spin susceptibility by inducing a resonance peak at  $(\pi/a, \pi/a)$  but also that it drastically changes its momentum dependences.

## **"Incommensurate peaks" in YBCO: not an evidence for dynamical stripes**

The observation of "incommensurate peaks" at some energy transfers [22, 23] has often been interpreted as clearcut evidence of dynamical stripes in YBCO. Our recent detailed study [18] basically rules out this conclusion (at least for near-optimally doped YBCO). Indeed, we established that the "incommensurate peaks" are only observed in the superconducting state and are additionally closely related to the commensurate resonance peak by a continuous dispersion relation (Eq. 1) as discussed above. This puts the observation of the magnetic incommensurability in YBCO in a totally new perspective. Being energy-dependent, temperature-dependent and doping-independent, the "discommensuration" is rather difficult to understand within a stripe picture where typically a characteristic distance (between charge stripes) needs to be observed. Without invoking any specific model, it becomes clear that their interpretation has to be necessarily related to the one made for the "commensurate" resonance peak.

The vanishing at  $T_C$  of the "incommensurate" excitations, we reported in YBCO<sub>6.85</sub> [18], can be actually anticipated over a wide part of the phase diagram [10]. [Notice that, even below  $T_C$ , it is still not established under which conditions and exactly in which doping range the "incommensurate" excitations are present in YBCO.] Nevertheless, their disappearance in the normal state is actually consistent with the different data published so far [22, 23, 28]. Indeed, the reports of normal state incommensurability in YBCO are rather scarce. At best, it is said that these incommensurate excitations remain in a small temperature window above  $T_C$  (up to 70-75 K for  $T_C=63$  K) [28] and finally disappear upon heating. But, as this intensity is weak on top of a phononic background (always present in such unpolarized neutron scattering experiments) whose structure factor mimics an incommensurate-like intensity modulation, no clear conclusion can be made and, at least, requires further work. In any case, fluctuations of the SC state (in the conventional meaning) could also explain the persistence of "incommensurate" excitations in a small temperature range above  $T_C$ . Recently, it has been argued that these incommensurate magnetic fluctuations have a one-dimensional nature [31]. This is based on measurements using a partially (half) detwinned YBCO<sub>6.6</sub> sample. Due to the above-mentioned phononic background and the scattering geometry used, this report is rather inconclusive: it is not proved that the observed effect is related to the magnetic scattering. Indeed, the detwinning of the sample can actually affect the background itself (for instance, if it is related to an  $a^*$ -polarized phonon). To make their point clear, these authors have to demonstrate that the balance of intensity between  $a^*$  and  $b^*$  is not present at high temperature (where the magnetic intensity is weaker and commensurate) or present polarized neutron beam data.

Our results in YBCO<sub>6.85</sub> [18] also contrast with those reported in the LSCO system [29, 30] where no change of the incommensurate peak position occurs across the superconducting temperature. In LSCO, the incommensurate peak structure begins to disappear only around room temperature [29]. Further, the observed energy range

where incommensurate peaks are observed is very different in the two systems (down to the lowest energies in LSCO but limited in a small energy range below  $E_r$  in YBCO). Their similarity is then reduced to only the symmetry of the incommensurate pattern along the (100) or (010) directions seen in both systems. However, the actual “fortified castle”-like shape observed in YBCO[22] looks rather different from the four well defined peaks observed in LSCO[30]. This makes dubious the universality of the spin fluctuations claimed to occur in the two systems[22] only based on the occurrence of “incommensurate” magnetic peaks. The origin of incommensurability in both systems likely requires a different scenario although common ingredients (such as Fermi surface topology) might be invoked.

As discussed above, our detailed study of the incommensurate magnetic peaks in YBCO shows that a standard interpretation within a ‘stripe phase’ picture is inconsistent. However, it should be noticed that a situation of strongly disordered stripes, as recently theoretically discussed in [32], is still possible. This would correspond to the case where the AF correlation length is lower than the mean distance between stripes [33]. And so, there is no  $\pi$ -phase shift from one AF cluster to the next one. These decorrelated AF clusters would give rise to the broad commensurate peaks observed in the normal state. However, the behavior of such objects in the superconducting state has not been addressed so far. This would be of great interest.

## Resonance peak and Superconducting condensation energy

The knowledge of the spin-spin correlation function in absolute units is becoming a crucial topic for the description of the physical properties of high- $T_C$  cuprates. For instance, magnetic neutron scattering has been recently proposed to provide a direct measurement of the condensate fraction of a superconductor[34]. A direct link with the high- $T_C$  mechanism has also addressed in the framework of the t-J model[35, 36]. The proposal is the following: if the SC pairing mechanism is due to AF exchange then the SC condensation energy,  $E_C$ , would be the energy gain between the normal state and the superconducting state of an exchange energy  $E_J$  of the form[35, 36]:

$$E_J = \frac{3J}{2\pi(g\mu_B)^2} \int_{BZ} d^2q [\cos(q_x a) + \cos(q_y a)] \int d\omega \frac{Im\chi(q, \omega)}{1 - \exp(-\hbar\omega/k_B T)} \quad (2)$$

where the sum over the wavevector is performed over the 2D Brillouin zone (BZ) and normalized by the BZ volume,  $(2\pi/a)^2$ . The condensation energy then reads,

$$E_C = E_J^{NS} - E_J^{SC} \quad (3)$$

It is essential to realize that Eq. 3 is a subtle net difference of the magnetic fluctuations spectral weight between the normal state and the superconducting state additionally **weighted** by a momentum form factor  $[\cos(q_x a) + \cos(q_y a)]$  corresponding to the Fourier transform of the AF exchange. It follows that the temperature dependent change in exchange energy crucially depends on a redistribution of the magnetic spectral weight in **momentum**. Indeed, according to Ref. [35] the exchange energy differs from the total moment sum rule,  $W = \int_{BZ} d^2q d\omega Im\chi(q, \omega)/(1 - \exp(-\hbar\omega/k_B T))$ , only by this momentum-dependent form factor. If one neglects this wavevector dependence in Eq. 2, Eq. 3 becomes meaningless as  $E_C$  will necessarily be zero to satisfy the sum-rule. The wavevector form factor in Eq. 2 is then essential and cannot be neglected.

In a recent Report, Dai et al. [6] have followed this idea and claim to have found a quantitative correspondence between the temperature derivative of the spectral weight

of spin excitations in YBCO and the electronic specific heat  $C_{el} \simeq dE_J/dT$ . We wish to point out that the analysis provided by Dai et al. fails at an elementary level as they fully neglected the wavevector form factor in Eq. 2 by rewriting  $E_J$  as,

$$E_J \simeq \frac{3J}{\pi(g\mu_B)^2} \int_{BZ} d^2q d\omega \frac{Im\chi^{res}(q, \omega)}{1 - \exp(-\hbar\omega/k_B T)} \quad (4)$$

where  $Im\chi^{res}(q, \omega)$  is only the resonant part of the spin excitations. Eq. 4 is derived by assuming that the spins accounting for the resonance part are fully decorrelated in the normal state in contrast with the observation of AF dynamical correlations above  $T_C$ . They then conclude that a large part of the electronic specific heat is due to spin fluctuations. There is no doubt that the electronic specific heat and the spin fluctuations are related in some way: after all, they are ultimately attributable to the same strongly interacting electron system. However, the analysis of Dai et al. [6] is much too crude to uncover this underlying relation. In an optimally doped sample, they finally obtain a contribution to the specific heat  $\sim$  **three** times larger than the measured one[37] (as found in [36]). In underdoped samples, the discrepancy is even bigger as the measured specific heat jump drastically falls down whereas the resonance peak spectral weight remains approximately constant for all doping[6, 9] as  $\int d^2q d\omega Im\chi^{res}(q, \omega) \sim 0.05 \pm .02 \mu_B^2$ , and so would be the calculated specific heat jump at  $T_C$ . [It should be noticed that this absolute unit value has been independently obtained by the two different groups]. Further, the attempt to relate the specific heat anomaly in the normal state with a speculated onset of the resonance peak at  $T^*$  (see above) is meaningless. Indeed, the most salient feature of the electronic specific heat [37] is its pronounced **increase** with increasing doping in the normal state. By contrast, the magnetic spectral weight strongly **decreases** with increasing doping in the same temperature range. These discrepancies do not necessarily suggest that the proposal of Eq. 2 is not correct. It just means that the analysis performed in Ref.[6], Eq. 4, relating the magnetic fluctuation spectrum and the electronic specific heat is invalid and inconclusive as it oversimplifies the physical content of Eqs. 2 and 3. As emphasized by Scalapino and White [35], the net difference in Eq. 3 will be very small and then difficult to estimate. To overcome this problem, Dai et al. [6] have arbitrarily considered **only** the contribution of the resonance peak spectral weight around  $(\pi/a, \pi/a)$  and at the energy  $E_r(Q_{AF})$  (that they attempt to relate to the electronic specific heat). The actual change of the spin susceptibility across the SC temperature as discussed above (dispersion behavior such as Eq.1) reveals that the estimate of Eq. 3 would be very subtle (especially in underdoped samples).

In conclusion, the resonance peak is certainly a key feature for the description of the physical properties of high- $T_C$  superconductors which has been widely reported at the commensurate AF wave vector. Further, the observation of its dispersion[18] experimentally suggests its collective nature. It now emerges that the role of such a magnetic collective mode would be essential for the interpretation of physical properties of high- $T_C$  superconductors, for instance, to describe the complex spectral structure of the one-particle spectrum[38] as reported by photoemission spectroscopy.

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Figure 1: Lower panel) Neutron intensity scans in the  $(H, K)$  momentum plane for a fixed energy transfer at  $E = 35$  meV (each scan has been shifted by 120 cnts from the lower one for presentation). The momentum transfer along  $c^*$  was fixed to the maximum of the magnetic structure factor  $L = 1.7[9]$ . The phonon background measured at room temperature has been subtracted from the data after proper correction of the temperature factor following a procedure detailed in [7]. Measurements have been performed on the 2T triple-axis spectrometer (Laboratoire Léon Brillouin, Saclay) with  $k_f = 2.662 \text{ \AA}^{-1}$ , the momentum resolution (FWHM) was 0.14 r.l.u. along  $H$  direction and 0.1 r.l.u. along  $K$  and the energy resolution was 4 meV. All scans have been fitted by either two Gaussians peaks displaced by  $\Delta H$  from  $H = 0.5$  or a single Gaussian peak centered at  $H = 0.5$ . Upper panel) Sketch of the reciprocal space around the AF wavevector. The squares represent the locus of maximum magnetic intensity in the superconducting state. The shaded area indicates the momentum  $(H, K)$  space covered by the q-scans of the lower panel.

